

Do these residues sent from Rockwell-Hanford at Richland, Washington contain salvagable plutonium? Members of LASL's Plutonium Facility determine the kinds and amounts of special nuclear materials in such heterogeneous waste with nondestructive assay techniques (the segmented gamma scanner and the thermal neutron coincidence counter). Measurements of this sample indicated that the contents were not worth recovering.

Nondestructive Assay for Nuclear Safeguards

by Roddy B. Walton and Howard O. Menlove

Sophisticated nondestructive assay techniques have solved many difficult measurement problems in the nuclear fuel cycle, but their potential for keeping track of sensitive nuclear materials on a nearly continual basis is just beginning to be exploited in nuclear facilities.

Radioactive nuclear materials reveal their presence by the radiation they produce spontaneously or otherwise. These characteristic signatures form the basis for nondestructive assay (NDA) of sensitive nuclear materials and for modern safeguards measurement technology. NDA techniques are used now by the nuclear industry, defense facilities, and safeguards inspectors to make rapid, accurate measurements of sensitive nuclear materials in diverse forms and compositions and thus to close many gaps in inventory measurements. However, this technology has not drastically changed the overall materials accounting practices of most nuclear facilities. Instead, the nuclear industry has placed heavy emphasis on physical protection to safeguard nuclear materials against overt threats of diversion. Further, a major part of the National Safeguards Program is devoted to increasing these

physical protection measures.

The Los Alamos Scientific Laboratory (LASL) program, on the other hand, has pioneered the development of NDA technology needed not only for inventory closure but also for near-real-time measurement and accountability systems. NDA techniques can be used to assure that no sensitive nuclear materials (SNM) are being lost or diverted from their defined flows and containments. Rapid measurements of SNM in feed materials, process lines, finished products, scrap and waste, and holdup in the plant are all possible with NDA techniques. When put together in an integrated materials control and accountability system, they can deter protracted diversion of SNM by a knowledgeable insider by detecting the amount and location of losses in a timely fashion. Such systems can also determine the validity of threats that significant amounts of nuclear material have

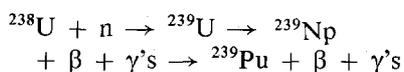
been diverted for unauthorized purposes.

At present several NDA-based near-real-time materials accounting systems are in existence and we are helping to plan such systems for future high throughput fuel production and spent-fuel reprocessing facilities. We also continue to work closely with members of the nuclear industry and safeguards inspectors to solve individual measurement problems associated with accurate inventory measurements, quality assurance of nuclear fuel production, and adequate safeguards inspection procedures. For many existing facilities, accounting systems that combine conventional chemical analysis with NDA techniques can provide adequate safeguards, but advanced facilities will require upgraded approaches.

Uranium and plutonium are the principal raw materials of fission weapons and hence the most sensitive nuclear materials to safeguard and to measure.



Moreover, because their fissile content or isotopic composition determines their strategic value as well as their value for nuclear reactor applications, it is important to measure isotopic compositions as well as absolute amounts of these elements. For example, natural uranium contains 0.7% of the fissile isotope ^{235}U , with the remainder being the fertile isotope ^{238}U . Both fissile and fertile isotopes will fission when bombarded with neutrons, but fissile isotopes are the important ingredient in nuclear fuels because they fission with high probability when bombarded by low-energy neutrons, whereas fertile isotopes will not fission unless they are bombarded by high-energy neutrons. Plutonium-239 is the principal fissile isotope in all plutonium fuels. This isotope is produced in nuclear reactors through the reaction



Uranium-238 is called fertile because it produces the fissile isotope ^{239}Pu by neutron capture. Other fertile isotopes are ^{240}Pu , which produces fissile ^{241}Pu by neutron capture, and ^{232}Th , which produces ^{233}U . Thus safeguards measurements are concerned with determining the presence of a large number of fissile and fertile isotopes of uranium, plutonium, and thorium.

In this article we discuss nuclear NDA methods based on detecting either the natural radioactivity of these materials (passive assay) or the radiations produced when these materials are bombarded by external sources of neutrons and gamma rays (active assay). The methods are rapid, usually requiring only a few minutes to complete a measurement. They are nondestructive in the sense that the materials can be measured without removing them from their containers and that the measurement is based on observation of the

decay or transmutation of a negligible number of nuclei relative to one mole. Moreover, they interrogate the entire bulk of the material rather than just a sample and are therefore capable of accurately measuring heterogeneous materials such as scrap and wastes from processing.

One of the earliest, most critical needs for NDA methods was for the measurement of nuclear process scrap and waste. Chemical analysis was not reliable because of the problem and expense of obtaining representative samples from these characteristically heterogeneous materials. Some large inventory discrepancies (for example, the problem encountered at the NUMEC facility in Apollo, Pennsylvania, in 1965) have been difficult to resolve because of deficiencies in such measurements. Good measurements of scrap also were needed to assess the material's value for recycle, and NDA methods for the screening and assay of low-level waste were needed to dispose of the materials safely and economically.

NDA techniques were also required to extend the ability of safeguards inspectors to check facility operator compliance with safeguards requirements—in particular, to check that measured material inventories matched declared "book" inventories within the limits of measurement uncertainties. To accomplish this objective, inspectors carry out detailed auditing of records and procedures; perform on-the-spot inventories of containers of nuclear materials, which involve weighing and verifying seals; collect random samples for chemical analysis; and examine analytical laboratory procedures. With portable and in-plant NDA instrumentation, together with certified standards, an inspector can independently verify the types and amounts of materials in a facility, including materials held up in process lines.

International Atomic Energy Agency

(IAEA) inspectors present special challenges for measurement instrumentation because they work under difficult political and physical constraints. The terms of the safeguards agreements between the country being inspected and the IAEA define the political constraints. For example, their terms limit materials accessibility and inspection time. Further, the inspector must work in a foreign environment without the normal supporting services, such as calibration standards, that are available for domestic safeguards. Equipment therefore must be lightweight, reliable, rugged, and easy to calibrate.

The LASL Safeguards Program has studied and continues to study these and other challenging measurement problems. In most cases, the problems have been identified by domestic and IAEA safeguards inspectors, Department of Energy (DOE) field offices, DOE facility operators, DOE safeguards systems analysts, and designers of integrated safeguards systems for new fuel cycle facilities, as well as the operators of LASL facilities, principally CMB Division staff. Because we want to gain widespread acceptance of our new technology, we consider test and evaluation of a fully engineered prototype instrument in the operating environment of a host nuclear facility to be the most important phase of the development of a measurement method. We also prepare comprehensive design and performance documentation to facilitate the work of instrumentation vendors and potential users.

In the remainder of this article we describe (a) techniques for nondestructive assay of fissionable material including some applications of major methods, (b) the status of NDA technology development and its implementation in the fuel cycle and DOE facilities, and (c) some future challenges in this field of measurement technology. Many laboratories over the world have

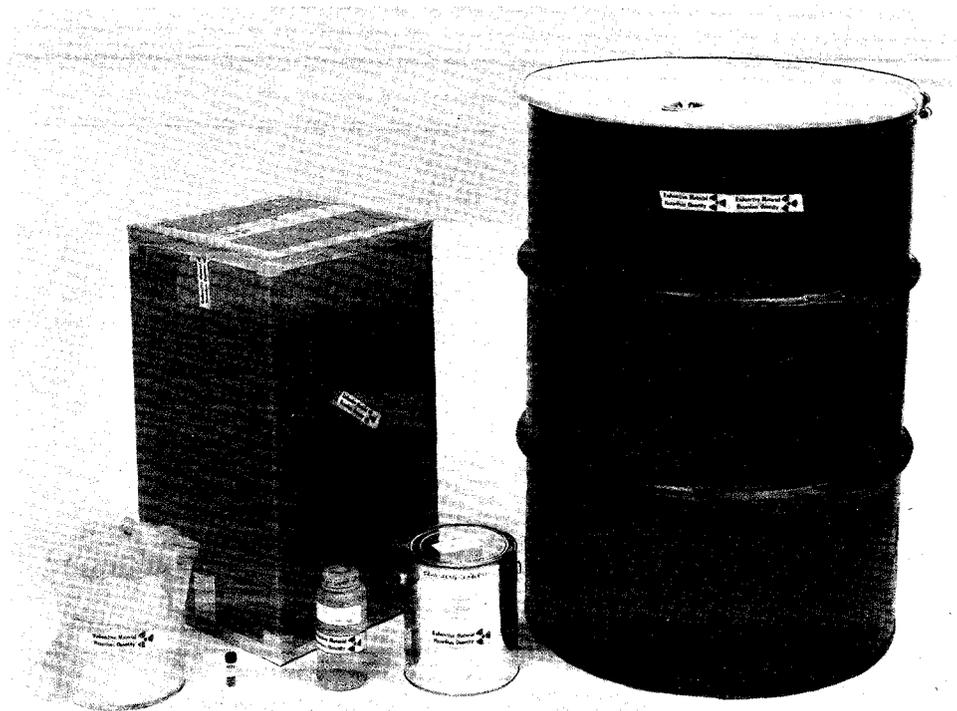


Fig. 1. Examples of containers used for nuclear materials in fuel cycle facilities. The large containers are used for low-concentration scrap and waste, the intermediate-size containers (1- and 4-L) hold high-purity process fuel materials or high-concentration scrap, and the small vial is a typical container for samples withdrawn from process lines for chemical analysis.

contributed significantly to the development of this technology, but rather than present a comprehensive review, we will focus on LASL-developed instrumentation and methods.

Characteristics of a Nondestructive Assay System

No single NDA instrument or method will suffice for the assay of the diverse forms and containments of nuclear materials in the fuel cycle. Some typical samples are shown in Fig. 1. Typical "feed" materials for input to the fuel fabrication process are UF_6 in metal cylinders with capacities for up to $\sim 10^4$ kg, depending on enrichment; plutonium nitrate solution in 10-L plastic bottles (120 cm long); and uniform fuel blends in few-liter cans. Reactor fuel materials include pellets, plates, rods, and bundles. Recoverable scrap, such as defective product material and calcined process residues, is stored in few-liter containers. Slightly contaminated wastes, such as paper, rags, and rubber gloves, are often stored in 120- and 220-L drums.

Clearly then, the capability to "view" bulk materials quantitatively is essential for most applications; hence, for both active and passive assay the interrogating beams and radiation signatures of the nuclear materials being measured must be highly penetrating. Neutrons and gamma rays are obvious choices from the domain of low-energy nuclear physics, as opposed to less penetrating alpha and beta rays. Additionally, calorimetry, which is based on the measurement of heat generated by radioactivity, can be used to measure plutonium with high precision, provided the isotopic composition of the plutonium is known from independent mass spectrometry or gamma-ray spectrometry measurements. (Uranium does not generate enough heat to be measured with calorimetry.)

The characteristics that are common to all practical NDA systems are:

1. an assay principle based on the detection of suitably copious fundamental signatures of the isotopes or elements to be measured, that is, neutrons or

gamma rays from natural radioactivity or from interrogation with an external source;

2. a detection system optimized for the signature selected and the types of samples to be measured;

3. electronic and mechanical systems for data acquisition and assay controls; and

4. a consistent means for obtaining the desired mass of the isotope or element from observed counting data.

Detectors for NDA instruments are essentially solids or gases that ionize as they interact with incoming neutrons or gamma rays. The amount of ionization is proportional to the energy deposited in the detector by each interaction. The generated charge forms an electronic pulse that is then amplified and processed and either counted or analyzed by electronic and microprocessor units. Gamma-ray detectors used in NDA instruments are solid crystals of NaI and germanium, standard tools of the gamma-ray spectroscopist. In NaI detectors, the ionization in the crystal produces light (scintillation), which is converted to an electronic pulse by a photomultiplier tube. In germanium and other solid-state detectors, the charge produced is converted directly into an electronic pulse. The neutron detectors most commonly used for NDA instruments are gas proportional counters, filled with 3He , BF_3 , or 4He ; however, plastic or liquid scintillators also can be used.

The conversion of counting-rate data to mass determinations or isotopic abundances of SNM involves the most subtle and difficult problems. Among these are calibration of the instruments and corrections for absorption and scattering of the signature radiation by the sample being measured. The sample includes not only the particular isotope or element of interest, but also "matrix" materials in which the material of in-

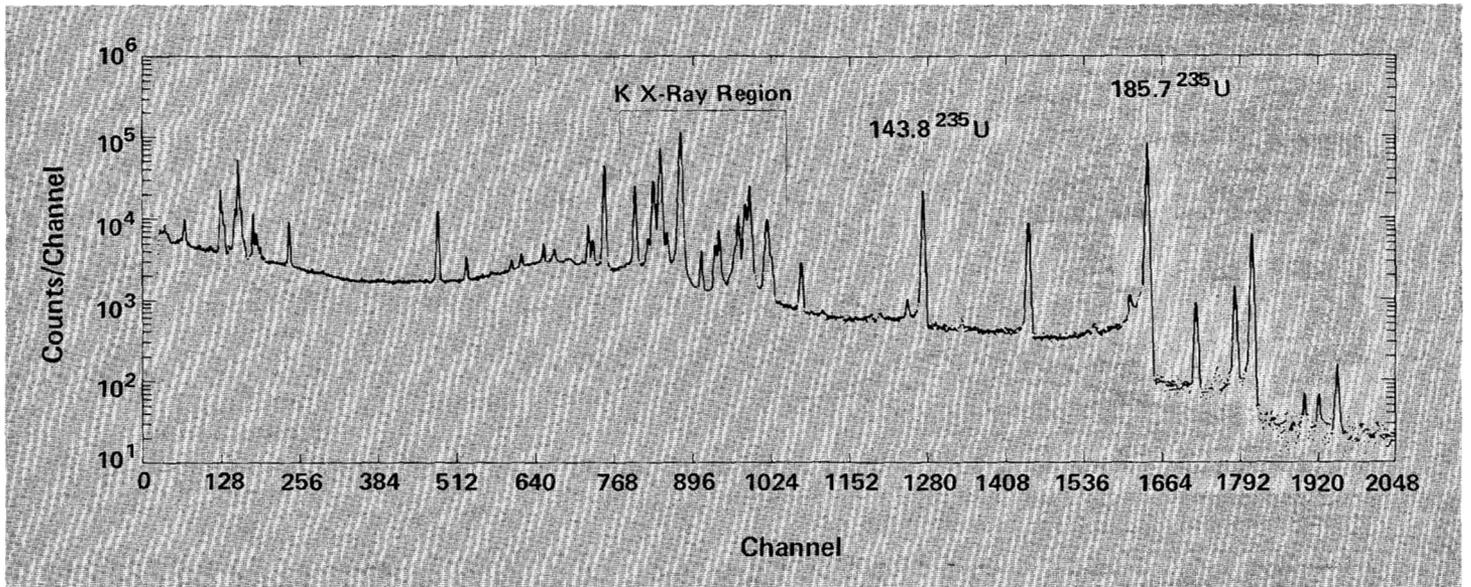


Fig. 2. Pulse-height distribution of gamma rays from high-enriched (93% ^{235}U) uranium, measured with a high-resolution germanium detector. Gamma-ray energies (in keV) are noted on some of the peaks. The prominent peak at 186 keV is used for passive assay of ^{235}U .

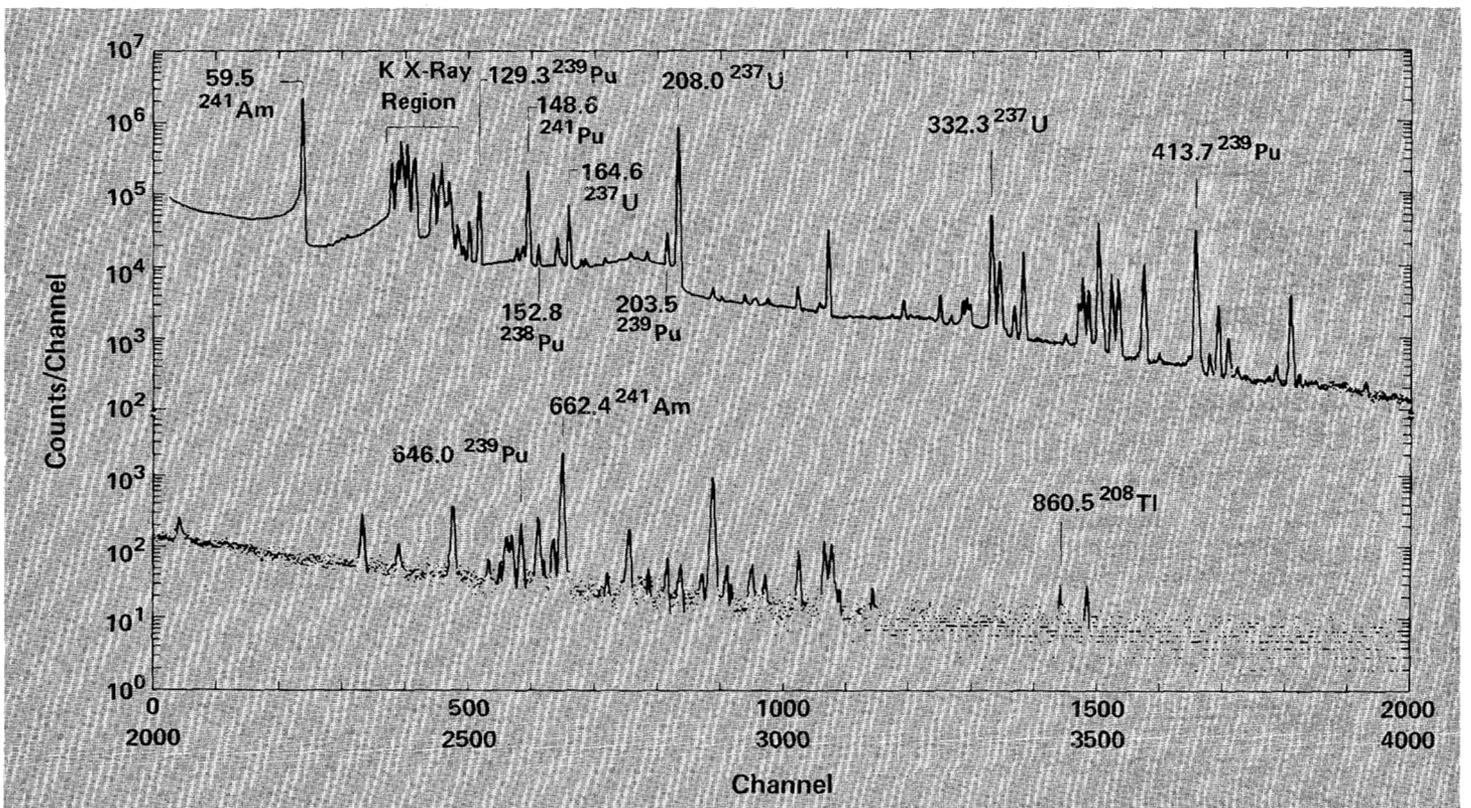


Fig. 3. Pulse-height distribution of reactor-grade plutonium (15% ^{240}Pu), measured with a high-resolution gamma-ray detector. Gamma-ray energies (in keV) are noted on some of the peaks.

terest is embedded. In an active method, one also has to account for the absorption and scattering of the interrogating beam of photons or neutrons as it penetrates the sample. If the chemical and isotopic compositions of samples are well known, as with identical cans of PuO_2 powder, then the correction problem may be circumvented simply by calibrating the measurements with a set of standards that closely cover the range of unknowns. However, we prefer a method whose response is as independent of sample matrix materials as possible, to reduce the number of calibration standards. It is even more important to reduce matrix dependence in measurements of poorly characterized scrap and waste materials. We use the sophisticated radiation transport codes developed over the years for defense projects and reactor design to guide the design of an assay system.

General Methods for Passive Gamma-Ray Assay

Each isotope of SNM is radioactive and decays at a known rate through alpha or beta decay (for example, $^{235}\text{U} \rightarrow ^{231}\text{Th} + \alpha + \gamma$'s and $^{239}\text{Pu} \rightarrow ^{235}\text{U} + \alpha + \gamma$'s). These spontaneous decays produce a characteristic spectrum of gamma rays, specific in both quantity and energy. Thus a direct measurement of the amount of SNM in a sample can be obtained by counting the gamma rays it emits at specific energies.

The pulse-height spectra of gamma rays from high-enriched (93% ^{235}U) uranium and from reactor-grade (15% ^{240}Pu) plutonium are shown in Figs. 2 and 3, respectively. The isotopic abundances of the reactor-grade plutonium are: $^{238}\text{Pu} = 0.15\%$, $^{239}\text{Pu} = 81.6\%$, $^{240}\text{Pu} = 15.2\%$, $^{241}\text{Pu} = 2.4\%$, $^{242}\text{Pu} = 0.66\%$, and $^{241}\text{Am} = 0.8\%$. These data were taken with a high-resolution germanium gamma-ray detector and a multichannel pulse-height analyzer. The

plutonium spectrum is very complex, with contributions from several hundred gamma rays from ^{238}Pu ; ^{239}Pu ; ^{240}Pu ; ^{241}Pu , and the daughters or products of ^{241}Pu decay; ^{237}U ; and ^{241}Am . (In Fig. 3, a few of the peaks are labelled with their energies and isotopic origins.) Furthermore, the gamma-ray spectrum of plutonium changes with time because of the relatively short half-lives of ^{241}Pu and ^{237}U .

An assay is based on determining the areas under one or more peaks in the spectrum, usually calculated with a minicomputer that is an integral part of a multichannel-analyzer, data-acquisition system. We use either NaI or germanium gamma-ray detectors, but we prefer germanium because its higher (30X) resolution permits cleaner separation and more accurate analysis of the gamma-ray spectrum peaks. On the other hand, for field measurements, such as the assessment of material holdup in process equipment, portable assay units comprising a NaI detector and single-channel electronic analyzers set to bracket specific peak and background regions of the gamma-ray spectrum are satisfactory.

For measurement of uranium, we use the prominent 186-keV gamma ray from ^{235}U ; 4×10^4 gamma rays are emitted per gram ^{235}U per second. Similarly we use the 414-keV gamma ray from ^{239}Pu , which has a comparable intensity, to measure plutonium. To convert counting rates of these isotopes to total uranium or plutonium, their isotopic abundances must be known or measured independently. In all applications except spent-fuel recovery, uranium isotopes other than ^{235}U and ^{238}U are present in such small amounts that they can be neglected. Uranium-238 can be measured using as the signature the 1.001-MeV gamma ray from the decay of its daughter $^{234\text{m}}\text{Pa}$. However, the measurement must be made at least 3 months after chemical purification of

uranium so that the daughters are in equilibrium. The isotopic abundances of plutonium are much more complex, but still they can be measured by performing a detailed analysis of gamma-ray pulse-height spectra (described below).

The simplest gamma-ray arrangement, a portable system that has found extensive application in plant surveys, is shown in Figs. 4 and 5. The assay system consists of a NaI detector housed in a lead collimator, pulse-processing electronics (contained in the small box in Fig. 5), a sample turntable, and an external gamma-ray source for determining the gamma-ray attenuation of the sample. An assay is performed by collecting counting-rate data with the unknown sample, measuring the transmission of an external beam of gamma rays through the sample, and applying a correction for the sample self-attenuation of the SNM gamma rays from the transmission measurement. We calibrate the system by performing measurements of known standards in the same manner.

The central problem in the NDA of bulk samples is the correction for sample self-attenuation. That is, the emitted gamma rays are scattered and absorbed within the sample itself. Attenuation is large and difficult to anticipate because the gamma rays have low energies, typically 100 to 400 keV; the samples frequently contain high-Z elements that absorb strongly; and the chemical composition of the sample is often unknown. The attenuation of gamma rays in a sample is given by $e^{-\mu\rho X}$ where ρ (g/cm^3) is the density of the medium, μ (cm^2/g) is the mass attenuation coefficient, and X (cm) is the thickness of the sample. The gamma-ray attenuation also can be written as $e^{-\mu_l X}$, where μ_l is the linear absorption coefficient. Table I gives the mean free path, $1/\mu_l$, of the ^{235}U 185.7-keV and the ^{239}Pu 413.7-keV gamma rays for several materials. Note that high-Z materials cause a dramatic

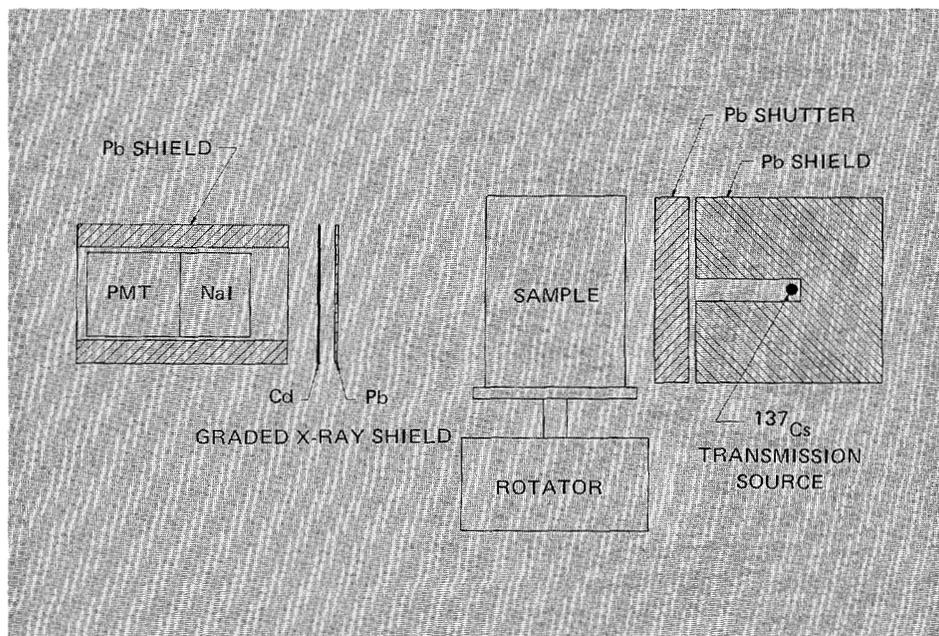


Fig. 4. A passive gamma-ray assay measurement setup. The sample is placed on a rotating table, and the detector (at the left) counts the gamma radiation from the sample. Correction for the attenuation of the SNM gamma rays by the sample is determined by measurement of transmitted gamma rays through the sample from an external radioactive source (in this case, ^{137}Cs) shown at the right.

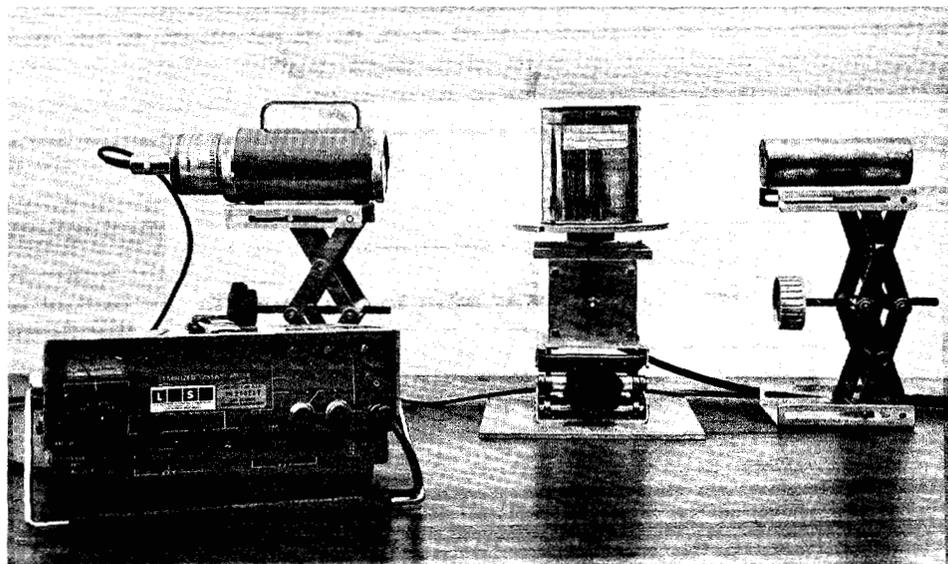


Fig. 5. A simple passive gamma-ray assay setup. At the left is the gamma-ray detector, a NaI crystal attached to a photomultiplier tube. The sample is mounted on a rotating table, and the transmission source is positioned to the right of the sample. (The arrangement is the same as that shown in Fig. 4.) The pulse processing electronics is an Eberline SAM-2 portable instrument located below the detector.